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FINAL REPORT

FOR

"COHERENT PROPAGATION AND SUM FREQUENCY GENERATION

INTO THE VACUUM ULTRA VIOLET"

AFOSR 81-0019

1 Nov 80 - 31 Oct 81

SUBMITTED BY

UNIVERSITY OF SOUTHERN CALIFORNIA

CENTER FOR LASER STUDIES

LOS ANGELES, CA 90007

Principle Investigator, Jean Claude Diels

I. INTRODUCTION.

The goal of this program is to demonstrate coherent sum frequency generation into the U. V., with energy conversion efficiencies of the order of several %. In the first year of this program, a facility was developed needed to perform these measurements. The source ~~that we constructed~~ consists in a synchronously pumped dye laser oscillator, tunable to the various two-photon transitions of lithium followed by a 3 stages Nd:YAG (frequency doubled) pumped dye laser amplifier. This facility will be moved to North Texas State University by the end of the first year of this program, where the actual measurements of UV generation will be made in the second year of this program.

The main technical discussion of the original proposal (which is reproduced below) still applies, except for the following points:

1. Most of the transitions of interest in lithium vapor (2s-2p, 2s-3d, 2s-3s) are in the laser emission band of the dye DCM. We demonstrated very efficient mode-locked operation of the synchronously pumped dye laser using DCM in benzol alcohol. Rh 6G should still be used for the 2s-4s transition.
2. A theoretical study was made of the influence of light coherence on two-photon resonant three photon ionisation of lithium. The number of ions produced is seen to be strongly dependent on the particular waveform of the excitation, in the case of coherent irradiation. This result should be exploited to minimize the ionization losses in our scheme of coherent sum frequency generation.
3. A revised cost estimate is included.

Errata

Pages 2 thru 4
Not Available

III. TECHNICAL DISCUSSION

III.1 Introduction

A review of the "state of the art" is given in Ref. 2 (Appendix B).

For the generation of vacuum ultraviolet (VUV) radiation, nonlinear crystals[3] and dye solutions[4] suffer catastrophic damage because of their ultraviolet absorption. Metal vapors and noble gases have the advantage of being nearly transparent in the region of the spectrum. While some photoionization will compete with the up conversion process, this process does not destroy the atom permanently.

The highest conversion efficiency (10%) was reported by Bloom et. al.[5] for nonresonant tripling of 1.06 μ radiation in a phase matched mixture of rubdium and Xenon. Conversion efficiencies of several % were achieved by other investigators[6,7] for the same system. Such a nonresonant scheme cannot yield high conversion efficiencies in the VUV because:

1. Competing photoionization - a four photon process at 1.06,μ, a two or three photon process for VUV generation
2. Weaker bound-free matrix elements for the visible wavelength - therefore, the incident intensity would have to exceed 10 GW/cm^2 over 1m distance, for optimum conversion, which is impossible.

A resonance condition can be sought to enhance the nonlinear susceptibility. Several experiments (listed in Ref. 1, Appendix A, Ref. 2, Appendix B) have been carried out to generate UV radiation

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by two photon resonant sum frequency generation. The conversion efficiency in this class of experiments has been low, because

a. Depletion of the fundamental by two-photon absorption
b. The linear susceptibilities are a function of the pump intensity, which varies with distance (pump depletion). Therefore, the "phase matching" of the fundamental and sum frequency cannot be achieved over long distances.

For instance, the highest intensity conversion efficiency (0.2%) was reported by Wallace and Zdasiuk[8] in a phase matched mixture of magnesium and Helium at an incident intensity of 0.5 GW/cm^2 with a 4 nsec pulse duration and output at 143 nm. The energy conversion efficiency (not reported) was at most 0.07%. The limitation a) and b) listed above are eliminated in the scheme proposed here:

a) using the reversibility of radiation - matter energy exchanges in conditions of coherent transient propagation to minimize the depletion of the fundamental;
b) by using properly phased two pulse sequences, such that the energy is transferred from one pulse (energy supply) into the next (harmonic generator) the pump intensity and thus the phase matching condition can be maintained constant over very long propagation distances.

Under these optimum excitation conditions, the efficiency of the sum generation process will depend on the relative position of the atomic levels with respect to the pump frequencies.

Guidelines are given in Ref. 1 (Appendix 1) to choose the best atom for a particular conversion scheme.

III.2 Principles of the Proposed Method

The theory of coherent TPR third harmonic generation is much more complicated than that of nonresonant THG.[5] In the latter case, the nonlinear medium is described only by a constant nonlinear susceptibility. However, in the case of a resonant interaction it is necessary to use the density matrix of the atomic system so as to be able to account for transient effects, population redistribution among the resonant levels and saturation effects. An additional complication is that the third harmonic photon generally exceeds the ionization limit of the atom (in the case of VUV generation in metal vapors).

All these effects have been taken into account in the theoretical model detailed in Ref. 1 and 2 (Appendix A and B). The reader is referred to the publication for the derivation of the equations and a rigorous analysis of the problem. A simple (qualitative) discussion of the main results is presented below, using the notations of Ref. (Appendix A).

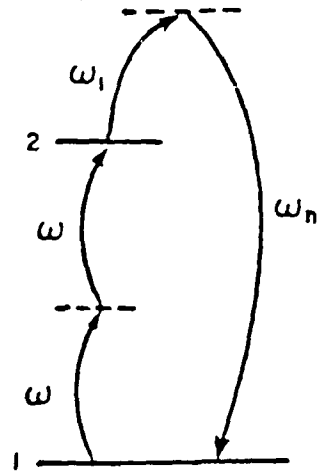


Fig. 1. Energy level diagram.

A strong pulse of frequency ω and amplitude $\mathcal{E} = \mathcal{E}(z,r)$ is resonant with a transition 1-2 (Fig. 1) A wave at ω_n (amplitude ω_n) is generated through the four wave mixing process $\omega + \omega + \omega_1 = \omega_n$. The

light field amplitudes are $E_i = \mathcal{E}_i e^{i(\omega t - kz)}$. The case of colinear (co-propagating) plane waves is considered here. The density matrix that describes the two photon excitation has an off diagonal element oscillating at 2ω, with amplitude σ_{12}. That off diagonal element can be viewed as an harmonic oscillation $Q = i\sigma_{12} e^{2i(\omega t - kz)}$, which will mix with the various fields to create the various nonlinear effects.

a) Two-Photon Absorption is described by the combination QE^* which represents a field at the pump frequency (ω) that opposes the fundamental field $E\left(\frac{\partial E}{\partial z} \simeq -QE^*\right)$

b) Sum Frequency Generation is described by the combination QE_1, which cumulatively (with distance) will increment the field generated at $\omega_n = 2\omega + \omega_1$ $\cdot\left(\frac{\partial E_n}{\partial z} \simeq - QE_1\right)$.

c) Feedback from the Generated Harmonic E_n into the Signal Wave E_1 results from the combination Q^*E_n $\cdot\left(\frac{\partial E_1}{\partial z} \simeq - Q^*E_n\right)$

The apparent limitation of resonantly enhanced harmonic generation can already be seen at this point. The amplitude of Q has to be large for maximum sum frequency generation (b), but then the two-photon absorption (a) is maximum as well. This is indeed the case in condition of "incoherent excitation" (pulse duration longer than the phase relaxation time of the medium and/or the pulse coherence time). In the conditions of two photon resonant coherent interaction, Q is no longer uniquely related to the field intensities, but has a complex (transient) evolution with time.

I have shown in an earlier publication[10] that it is possible to reverse the sign of Q during a portion of the signals whereby the two photon absorption (a) is replaced by two-photon stimulated emission. In order to see how this occurs, we need only consider the relevant part of one of the equations of motion of the density matrix (a of the oscillation Q):

$$\dot{Q} = - WE^2$$

Here, $W = \sigma_{22} - \sigma_{11}$ is the population difference of the two level system. Initially, $Q = 0$ and $W = -1$. Q can only be made negative if, during a portion of the pulse:

a) $W < o$ the medium has been driven by a short pulse into an inverted state[10]

b) $E^2 \rightarrow -E^2$ the phase of the exciting pulse has been shifted by 90^o, thereby reversing the sign of the driving term E^2. .

The first method requires very intense pulses. Because of transient Stark shifts associated with the high intensities, the oscillation Q and the generated signal have strong phase and amplitude modulation. The conversion efficiency per unit bandwidth is low. The second method was shown[1,2] to yield the highest conversion efficiency through an appropriate choice of the pulse parameters, and the phase matching condition. The two photon stimulated emission that occurs in the second half of the signal can be made to compensate the other losses (in particular photoionization). The (nonequilibrium) susceptibilities at ω, ω_1 and ω_n are therefore conserved as that portion

of the pulse propagates through the medium and phase matching is possible over long propagation distances. The "pulse shapes" required to generate the phase shifted signal is simply an interferometric delay line. The field intensity transmitted by the delay line is

$E = \mathcal{E}(t)e^{i\omega t} + \mathcal{E}(t + \tau)e^{i(\omega t+\omega\partial}$ which, for $\omega = (2N+1)\pi/2$,

becomes $E = \mathcal{E}(t) + \mathcal{E}(t + \tau)e^{i\frac{\pi}{2}} e^{i\omega t}$

Maximum Achievable Conversion Efficiency

For a given atomic system, the maximum achievable conversion efficiency is of the order of the ratio η of the probabilities of the Raman channel $(\omega_1, -\omega_n)$ to the two photon process $(\omega, +\omega)$. This number η is equal to 0.1 for third harmonic generation $(\omega_n = 3\omega)$ through the 2s - 4s resonance of litnium. As shown in Ref. 1 (Appendix 1), this efficiency can be greatly enhanced if a near resonance condition can be found at ω_r (in addition to the two-photon resonance).

III.3 EXPERIMENTAL CONSIDERATIONS

The main requirements for the experimental demonstration of two photon resonant coherent harmonic generation are

1. A tunable picosecond source, of sufficient energy per pulse to be able to completely invert a two photon transition
2. A pulse shaper to generate the phase shifted pulses
3. A heat pipe oven to provide an homogeneous mixture of lithium and magnesium.

III.3.1 The Picosecond Source

The source requirements for the proposed two photon coherent interaction experiment are the same as for single photon self-induced transparency, except for the higher energy density required. The pulse should be bandwidth limited, single transverse mode. In the case of single photon coherent propagation the requirement of bandwidth limited pulse could be waived to some extent, because self-induced transparency can act itself as an ideal filter.[11,12,13] In the case of two photon coherent propagation, however, there is no "slow pulse" which can ignore part of the frequency content of the input pulse.[13] The "lossless" (quasi steady state) pulses are unstable, and the transmitted signal would always be dependent upon the modulation of the initial pulse.[10]

There are four possible choices (see Figure ?) of two photon transitions in Li that are accessible to the tuning range of efficient synchronously mode-locked cw dye lasers. These are the 2s-3s (7351 Å), 2s-4s (5712 Å) 2s-3d (6393 Å),

and the 2s-4d (5461 Å) transitions. The calculations presented in Ref. 1 and 2 pertained for the 2s-4s transition, whose wavelength (5712 Å) is near the peak of the gain of the Rhodamine 66 dye (laser), i.e., at a spectral location where stable and highly reproducible picosecond pulses can be easily obtained. The 2s-3d transition would require nearly an order of magnitude lower laser power densities (for comparable excitation), because of the proximity of the 2s-2p (6710 Å) single-photon resonance, and is consequently another principal resonance of interest in the present study. The 2s-4d is also of special interest, because it is associated with a much smaller Stark-shift than the other transitions

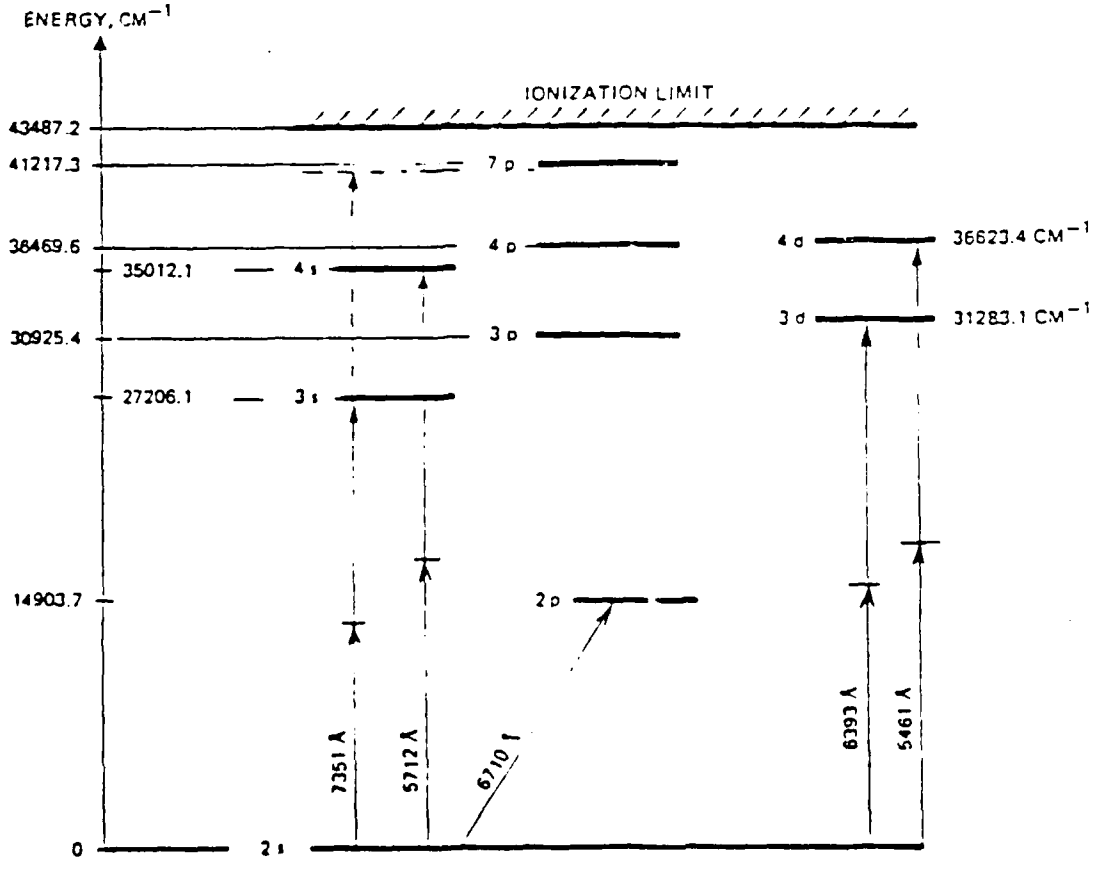


Figure 2. Energy level diagram of lithium (based on data from Ref. 14)

Synchronously mode-locked dye lasers pumped by mode-locked ion lasers have recently been demonstrated to yield stable and highly reproducible picosecond pulses of adjustable duration over broad tuning ranges.[15,19] With an appropriate choice of dyes, and pump wavelengths from mode-locked ion lasers, picosecond pulses should be obtainable from appropriate dyes at all of the transitions of interest in the present work. These are listed in Table 1 with the corresponding excitation wavelengths, candidate dyes and corresponding pump lasers.

Table 1. Excitation Wavelengths and Choice of Dye Lasers for Lithium Transitions

Transition	Excitation Wavelength, Å	Dye Laser[a]	Pump Laser	
			Wavelength	Source
Single-Photon:				
2s → 2p	6709.6	Rh.101/Cr.V	5145 Å 5700 Å	Ar^+ Rh.6G
Two-Photon:				
2s → 3s	7351.3	Nile Blue	6741 Å	Kr^+
2s → 4s	5712.0	Rh.6G/Rh.110	5145 Å	Ar^+
2s → 3d	6393.2	Rh.B	5145 Å	Ar^+
2s → 4d	5461.0	Sodium Fluorescein	4880 Å	Ar^+

[a] Rh. ≡ Rhodamine; Cr.V ≡ Cresyl Violet

The peak power of the available synchronously pumped dye laser is only in the 10kW range. Doubled Nd-YAG pumped dye amplifiers will be used to amplify these pulses to the 100 MW range. Therefore a doubled Nd-YAG laser is requested for this program.

III.3.2 Generation of Phase Shifted Pulses - Pulse Shape Measurements

A 90^{o} pulse can be generated by splitting a pulse in two beams, and recombining it after delaying one path a quarter number of half wavelengths. A prototype of such an interferometer has been developed by the author.[20,22] This rugged construction (Figure 3) requires only a single mechanical adjustment. The optical paths are sketched in Figure 3. Frustrated total reflection is used to split the beam into two parts. The fractional transmission of the beam splitters is controlled by adjusting the pressure on the prisms as indicated by the arrows. One of the beams propagates through fixed glass prisms. The other beam comes out of the glass structure, and is reflected back by a movable prism (the position of which determines the time difference between the two lobes of the recombined signal). By tuning the pressure of the gas in the extended arm of this interferometer, it is possible to make adjustments of the delay on the order of fractions of optical wavelengths. It was found possible to adjust this Michelson type interferometer for complete extinction of a beam of 10 mm diameter.

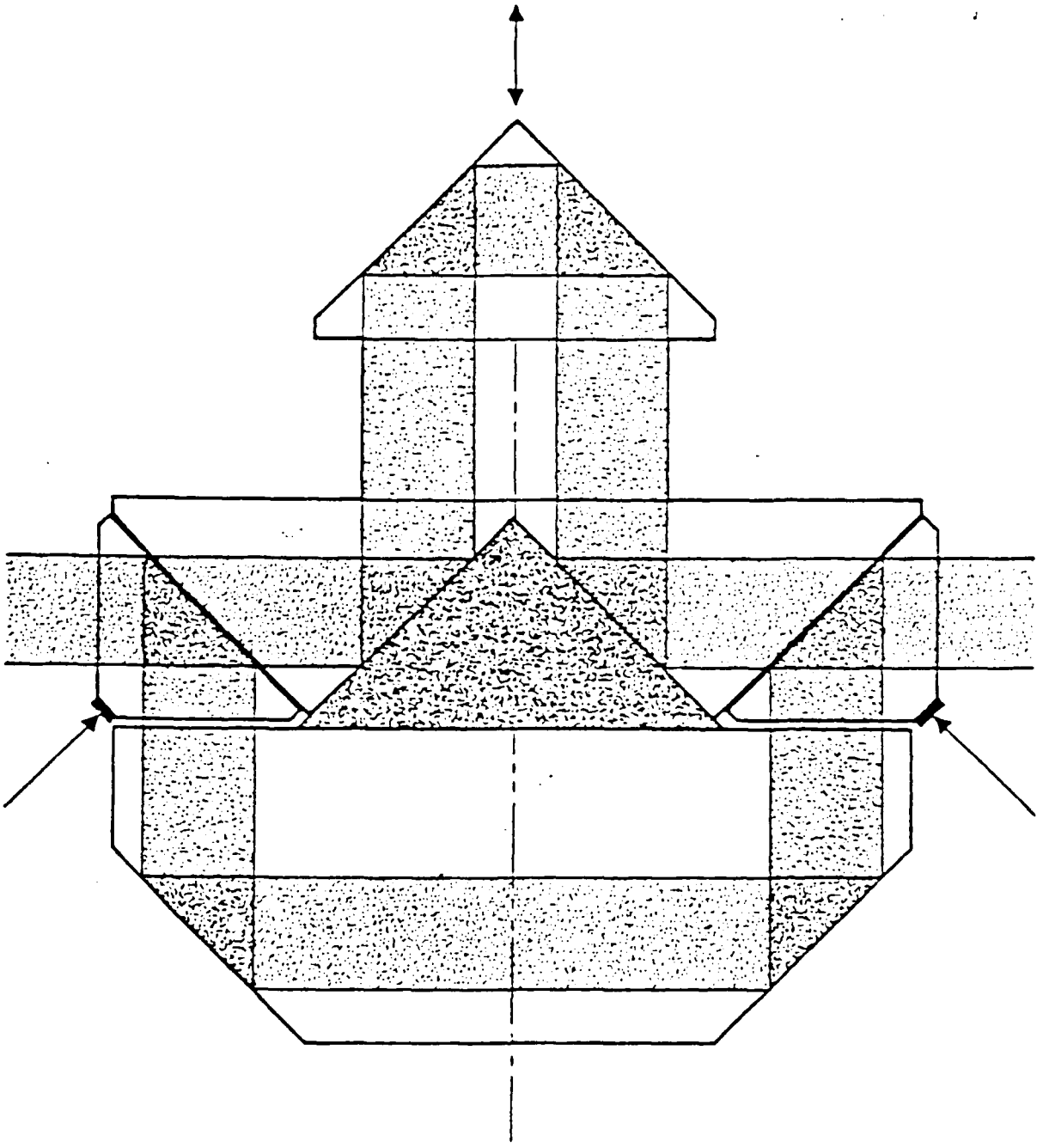


Figure 3. Design and optical layout of the rugged Michelson-type interferometer.

The theoretical investigation reported in Appendix A and B clearly demonstrates the need for a precise knowledge of the pulse shape, in amplitude and phase. When used for pulse duration measurements, this compact instrument has a resolution of 0.03 ps[20] and provides in a single measurement both linear and nonlinear autocorrelation functions.

Autocorrelation measurements with a peak to background ratio of 8 to 1 were recorded for the first time with this instrument.[23] This type of measurements were shown to yield phase as well as amplitude information[23], in contrast to the conventional autocorrelation measurements where all phase information is lost.

The autocorrelator just described will be used for measurements of the shape of the pulse incident on the heat pipe, as well as for generation of phase shifted pulses.

A 90° phase shifted pulse for instance is obtained by mechanically setting the delay to one half pulse duration, and fine adjustment of the gas pressure to have an optical path difference of $(2N + 1)\ \lambda/4$ at the beam recombination interface. One output beam of the autocorrelator will carry a $+90^{\circ}$ phase shifted pulse, the other a -90° phase shifted pulse.

Autocorrelation measurements on the pulses transmitted by the heat pipe will require a second autocorrelator, which we propose to construct in this program

Spectral measurements of the third harmonic will be made to compare theory and experiment. These spectral measurements alone will not provide a complete information on the pulse shape in amplitude in phase. The autocorrelator available now cannot be used for this purpose, because the prisms are made of BK7 glass, which is not transparent at the third harmonic wavelength. We propose therefore to construct the new "autocorrelator" in Suprasil (UV grade fused silica). Complementary information to the pulse spectrum will be collected from two different measurements: 1) straight second order autocorrelation using two photon photoionization detection; 2) cross correlation of the fundamental with the third harmonic. In the first technique the third harmonic is sent through the UV autocorrelator into a cell containing O_2 at low pressure. The ionization threshold of O_2 being 102.6 nm, it requires two photons at the third harmonic frequency to be ionized. The ionic charge (produced by two photon ionization of O_2 through the UV pulse at 192 nm) will be measured as a function of the delay setting τ_D of the autocorrelator, yielding the desired autocorrelation function. Cross correlation of the fundamental with the third harmonic is obtained by sending the output of the heat pipe (including fundamental and third harmonic) directly through the "interferometric autocorrelator" (now a "cross correlator"), into a KDP crystal to generate the frequency difference $3\omega - \omega = 2\omega$.

Measurement of the light energy generated at 2ω, as a function of the delay setting, τ_D of the interferometer, gives the cross correlation function:

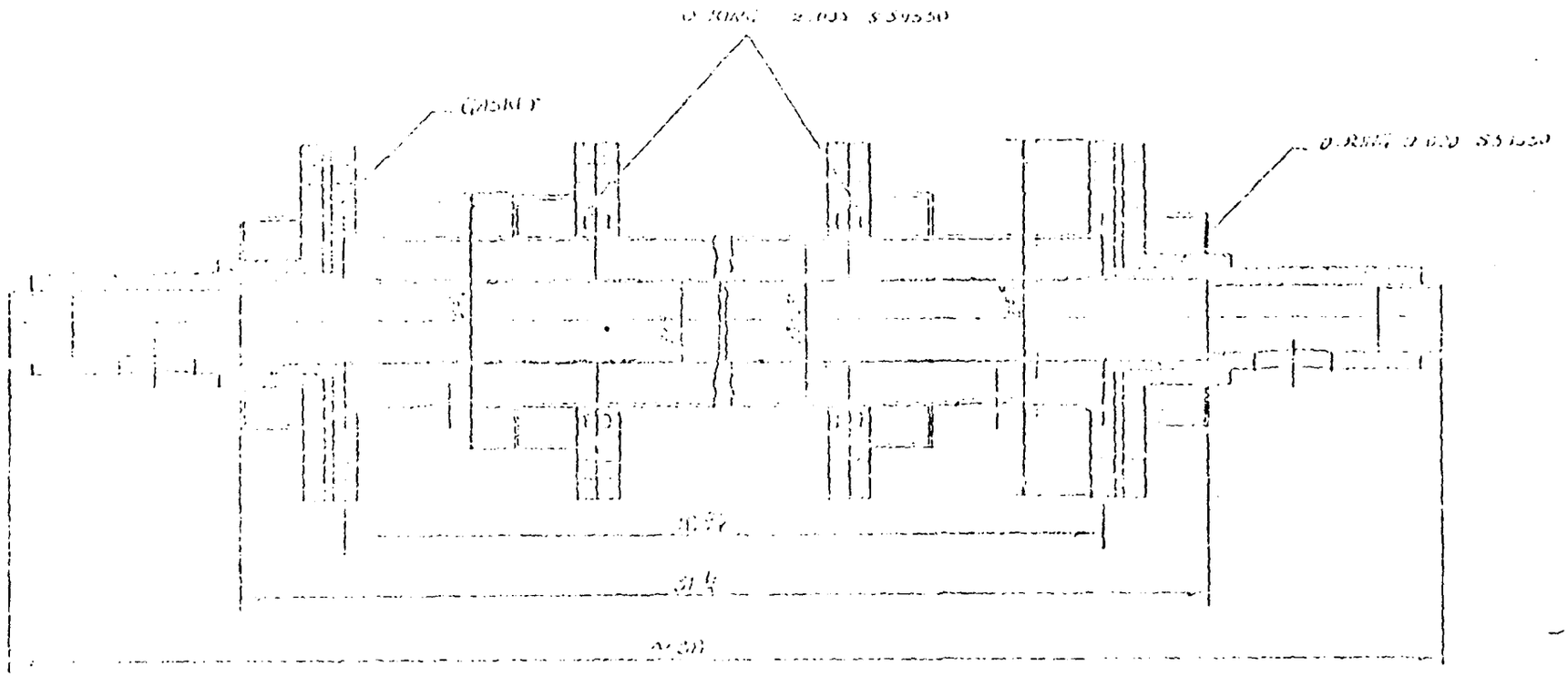
$$G_{2\omega}(\tau) = \int | \mathcal{E}_3(t)\ \mathcal{E}_1(t - \tau_D) |^2\ dt$$

This measurement in particular will provide knowledge of the relative phase of the fundamental and third harmonic, information essential for parametric amplification of the third harmonic.

III.3.3 The Lithium Magnesium Heat Pipe

In order to make a quantitative comparison with our theory possible, it is essential to have an homogeneous resonant medium at a well defined pressure and temperature. The first "heat pipe oven" developed for spectroscopic measurements by Vidal and Cooper[24] was precisely designed for lithium. A concentric heat pipe oven proposed and demonstrated by Vidal and Heller[25] provides homogeneous metal vapor inert gas mixtures as for instance, lithium-krypton. A slightly modified version, as proposed by Vidal and Hessel[26] was designed and built in our laboratory for mixing two metal vapors as lithium and magnesium. (Fig. 4) Our heat pipe has an active length of 50 cm, and is made of a SS347 for corrosion resistance in the presence of lithium. This heat pipe will be available for this project, and is adequate for the first part of the proposed program. A second heat pipe will have to be constructed for the parametric amplification measurements involving two heat pipes.

Figure 4.



HEAT PIPE OVEN
Scale 1:1.